APPENDIX D

EVALUATION OF A 10 MILLIREM PER YEAR BASIC RADIATION DOSE LIMIT FOR SETTING SOIL GUIDELINES AND SLUDGE LIMITS FOR RADIONUCLIDES

RESULTS OF RESRAD MODELING FOR RADIONUCLIDES IN LAND APPLICATION SITE SOILS

EVALUATION OF A 10 MILLIREM PER YEAR BASIC RADIATION DOSE LIMIT FOR SETTING SOIL GUIDELINES AND SLUDGE LIMITS FOR RADIONUCLIDES

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Prepared by E.A. Stetar, CHP

Performance Technology Group, Inc.

Nashville, Tennessee

INTRODUCTION

The RESRAD 6.0 code—developed by the Environmental Assessment Division of Argonne National Laboratory—was used to develop concentration guidelines for radionuclides in soils on the land application sites located on the Department of Energy (DOE) Oak Ridge Reservation (ORR). These application sites are used for the disposal of municipal sludge from the City of Oak Ridge Publicly Owned Treatment Works (POTW). The RESRAD code has been used extensively for assessments of potential doses form residual radioactive contamination in soils for both Department of Energy (DOE) controlled and Nuclear Regulatory Commission (NRC) licensed sites. The code calculates doses to a hypothetical resident farmer who lives on the contaminated site and obtains significant portions of his food and water from the site. Version 6.0 of RESRAD allows both deterministic and probabilistic dose analysis and risk assessment.

A basic radiation dose limit of 10 mrem/year was used as the basis for the land-application site soil guidelines. The 10 mrem/year limit was chosen because it is the value currently used by the City of Oak Ridge to develop limits for radionuclides in the POTW sludges. The City previously used a 4 mrem/year limit, but found it necessary to increase the dose limit, and corresponding sludge limits, to allow for future industrial growth.

The City currently has sludge limits for uranium, ^{60}Co, ^{134}Cs, ^{137}Cs, ^{154}Eu, ^{152}Eu, ^{54}Mn, ^{90}Sr, and ^{65}Zn. These limits are based on soil guidelines that were calculated using the RESRAD code with a basic radiation dose limit of 10 mrem/year (Stetar 2000). The soil guidelines are converted to sludge limits based on the expected dilution (i.e., mixing of sludge into soil following application) and the amount of loss to radioactive decay that will occur between the time of application and the time the site becomes available for unrestricted use. The City uses its sludge limits as a basis for determining the maximum quantities of the various radionuclide that can be discharged to the Oak Ridge sewer system each month. These total acceptable discharge quantities, called allowable loadings, are allocated among the radioactive materials dischargers on the basis of need. The allocations are accomplished through the City's industrial pretreatment permitting program. It should be noted, that because some of the permitted industries discharge only occasionally and because those that do discharge more frequently rarely discharge the entire allocated quantity, the radionuclide levels in the Oak Ridge sludge are at concentrations well below the calculated acceptable limits for sludge.

However, because the original allowable loadings—calculated on the basis of a 4 mrem dose limit—had for the most part been completely allocated, the decision was made to update the program using a somewhat less conservative dose limit of 10 mrem/year. The soil guidelines and sludge limits calculated using the higher dose limit result in larger allowable loadings for the key radionuclides entering the Oak Ridge treatment plant. Therefore, the updated limits allow the program to expand to meet the needs of future growth. Although the allowable loadings have been increased, the City of Oak Ridge does not anticipate that the concentrations of radionuclides in its sludge will actually exceed the limits originally established using the 4 mrem dose limit. It is the City's intention to maintain the radionuclide levels in the land application site soils to levels that are as low as reasonably achievable (ALARA). Oak Ridge has established a daily radionuclide screening program for sludge. Furthermore, in depth analyses are performed on the sludge and land application site soils on a regular basis. If analyses indicate any significant increase in radionuclide levels, the City could modify its land application procedures (primarily loading rates) to ensure soil concentrations are kept at a minimum.

It should be noted that the 10 mrem/year dose limit used for setting sludge limits is one-tenth the dose limit established by the NRC for members of the public when exposed to radiation from NRC or State-licensed facilities. Furthermore, it is less that half the dose limit established by the NRC—Final Rule on Radiological Criteria for License Termination—for use in deriving radiological criteria for unrestricted use following decontamination and decommissioning of licensed facilities.

PURPOSE

Soil guidelines and sludge limits—based on a 4 mrem/year dose limit—were previously calculated for 23 radionuclides in *Environmental Assessment, Proposed Changes to the Sanitary Sludge Land Application Program on the Oak Ridge Reservation* (DOE 1996). These guidelines and limits are provided in ***Table D.1.***

For this work, seven of the radionuclides from ***Table D.1.*** (^{60}Co, ^{137}Cs, ^{152}Eu, ^{155}Eu, ^{234}U, ^{235}U, and ^{238}U) and five additional radionuclides, ^{54}Mn, ^{65}Zn, ^{90}Sr, ^{134}Cs, and ^{154}Eu, are addressed. These radionuclides were chosen either because they have been detected in the Oak Ridge sludge or are believed to have a high potential for discharge to the Oak Ridge POTW. For the radionuclides listed in ***Table D.1.*** that are not addressed here, the previous soil guidelines from DOE/EA-1042 will continue to apply.

For these 12 primary radionuclides of interest, the RESRAD 6.0 code was used to calculate soil guidelines on the basis of a 10 mrem/year basic radiation dose limit to ensure consistency with the City of Oak Ridge's sludge management program. RESRAD 6.0 was also used to estimate the potential risks to the hypothetical resident farmer who establishes residency immediately following the last application of sludge. The risks that were estimated are the risk of cancer incidence (both fatal and nonfatal). One goal of this work was to ensure that the risk estimates for the soil guidelines do not exceed 10^{-4} (1 in 10,000). This risk level is considered appropriate because it is the "risk target" used by the Environmental Protection Agency (EPA) to establish regulatory limits for carcinogens in land applied sludges. Radionuclides are not currently addressed in the EPA's sludge standards.

Because the exposure scenario used in this work (the resident farmer) is highly conservative, the risk estimates reported should be considered upper bounds. The risk estimates associated with more probable exposure scenarios are provided in the human health risk assessment performed as part of this EA. For the seven radionuclides addressed in the previous EA (DOE/EA-1042), a comparison is provided of the risks associated with the former soil guideline values (based on 4 mrem/year) and those calculated here.

In addition to the calculation of soil guidelines and sludge limits, a probabilistic assessment was performed using RESRAD 6.0, to estimate the uncertainties associated with the dose and risk estimates for the radionuclides of interest.

Calculation of Soil Guidelines

To calculate soil guidelines for the application sites, the RESRAD code was initially run with the input soil concentrations for each of the 12 radionuclides of interest set to 1 pCi/g and the basic radiation dose limit set to 10 mrem/year.

Table D.1. Soil Guidelines and sludge limits as reported in *Environmental Assessment, Proposed Changes to the Sanitary Sludge Land Application Program on the Oak Ridge Reservation,* DOE/EA-1042. Final. October 1996.[a]

Radionuclide	Soil Guideline ($pCi/g_{\text{-dry wt.}}$)	Sludge Limit ($pCi/g_{\text{-drywt.}}$)
^{227}Ac	0.56	12.2
^{241}Am	7.7	167
^{60}Co	0.49	10.7
^{137}Cs	2.0	43.6
^{152}Eu	1.1	24.0
^{155}Eu	1.0	21.8
^{152}Gd	19.6	427
^{3}H	520	11,324
^{40}K	5.5	120
^{237}Np	1.5	32.7
^{231}Pa	0.81	17.6
^{210}Pb	2.5	54.4
^{238}Pu	9.1	198
^{239}Pu	8.3	181
^{226}Ra	0.11	2.4
^{228}Ra	0.95	20.7
^{99}Tc	35.5	773
^{228}Th	0.66	14.4
^{229}Th	1.5	32.7
^{230}Th	14.8	322
^{233}U	30.2	658
^{234}U	31.0	675
^{235}U	7.2	157
^{238}U	21.1	459.5

[a] taken from Table 1, page D-43 of DOE/EA-1042 (DOE 1996).

The radionuclides of interest were identified based on previous monitoring of the Oak Ridge sludge and information contained in the pretreatment questionnaires submitted to the City of Oak Ridge by all potential dischargers of radioactive materials. Short-lived medical radionuclides, such as I-131, were not included because their short half-lives would preclude significant build up on the land application sites. For the soil guideline calculations, the depth of the contaminated zone parameter was set to 0.15 m, and the area of contaminated zone parameter was set to 200,000 m^2. Previous studies conducted on the ORR land application sites indicate that a significant portion of applied radionuclides remain within the top 0.15 m of soil (Smith 1997, Boston et al., 1990). Furthermore, 0.15 m is the plow-layer depth used by the EPA in development of standards for metals and other contaminants in land applied sludges. The contaminated zone area of 500,000 m^2 corresponds to field size of approximately 120 acres. The current application sites range in size from approximately 25 to 117 acres.

Adjustment of Soil Guidelines

Uncertainty analyses were performed on the initial soil guideline values. The purpose was to identify any radionuclides for which further refinement in the modeling was needed to ensure the final calculated soil guidelines are sufficiently conservative (i.e., that the risk estimates for each radionuclide do not exceed 10^{-4}). Using information provided in NUREG/CR-6676, *Probabilistic Dose Analysis Using Parameter Distributions Developed for RESRAD and RESRAD-BUILD Codes* (Kamboj et al., 2000), the most sensitive parameters were identified for the radionuclides of interest (***Table D.2.***). The RESRAD code was used to perform the uncertainty analysis which consisted of a probabilistic assessment in which distributions (rather than point estimates) were used to represent the most sensitive parameters. For the parameters listed in ***Table D.2.*** the RESRAD default distributions were used and for the "area of contaminated zone" parameter a uniform distribution ranging from 40,470 m^2 to 607,050 m^2 was assumed. The contaminated area range represents potential application site sizes from 10 to 150 acres.

For those radionuclide for which the uncertainty analysis indicated maximum risks estimates greater than 10^{-4}, adjustments were made in the most critical model parameters, and the code was rerun to calculate a new soil guideline concentration. This process was repeated until a soil guideline value was obtained for which the maximum risk estimates did not exceed 10^{-4}.

The initial maximum risk estimates for ^{90}Sr and ^{65}Zn were on the order of 10^{-3}. The most critical parameters for these radionuclides were found to be the plant transfer factor for strontium and the plant transfer factor and distribution coefficient for zinc. For ^{90}Sr, a more conservative plant transfer factor of 0.95 was used in lieu of the RESRAD default of 0.3. The value selected represents the average of the transfer factors reported for clay/loam soils by the International Atomic Energy Agency (IAEA 1994). In the case of ^{65}Zn, the RESRAD default distribution coefficient of 0 was replaced with a more conservative value of 1800 cm^3/g, the average of the values for loam, clay, and organics, reported by the IAEA. The plant transfer factor for zinc was raised to 1.5 from the default of 0.4 based on the IAEA values (IAEA 1994).

Table D.2. Most Sensitive Parameters and Dominant Pathways for Radionuclides of Interest

Nuclide	Dominant Pathway	Most Sensitive Parameters			
^{54}Mn	external	SHF1	BRTF(1)	DROOT	DCACTC
^{60}Co	external	SHF1	DCACTC	BRTF(1)	BRTF(2)
^{65}Zn	external	SHF1	BRTF(1)	DCACTC	DROOT
^{90}Sr	plant	BRTF(1)	DROOT	DCACTC	BRTC(2)
^{134}Cs	external	SHF1	BRTF(1)	DCACTC	BRTF(2)
^{137}Cs	external	SHF1	BRTF(1)	DCACTC	BRTF(2)
^{152}Eu	external	SHF1	DCACTC		
^{154}Eu	external	SHF1	DCACTC		
^{155}Eu	external	SHF1	DCACTC		
^{234}U	plant	BRTF(1)	DROOT	DM	DCACTCU
^{235}U	external	SHF1	DCACTC	BRTF(1)	
^{238}U	ext & plant	SHF1	BRTF(1)	DROOT	DCACTC

SHF1=external gamma shielding factor; BRTF(1)=transfer factor for plants; BRTF(2)=transfer factor for meat; DROOT=depth of roots; DM=depth of soil mixing; DCACTC=distribution coefficient-contaminated zone.

Risk Estimates

Once the final soil guidelines were established, the RESRAD code was used to make estimates of the risk to the hypothetical resident farmer who moves onto the site immediately following the last application of sludge. RESRAD uses the EPA slope factors from the 1992 Health Effects Assessment Summary Tables (HEAST). However, for this assessment the RESRAD default coefficients for the radionuclides of interest and their progeny were replaced with the more recent risk coefficients found in Federal Guidance Report 13 (Eckerman 1999). The risk factors that were used are the morbidity risk coefficients for inhalation, ingestion, and external exposure. These coefficients estimate the risk to an average member of the U.S. population—per unit activity inhaled or ingested for internal exposures or per unit time-integrated activity concentration in soil for external exposures—of experiencing a radiogenic cancer as a result of intake of the radionuclide or external exposure to its emitted radiations.

For inhalation exposures, the absorption type for the particulate aerosols was assumed to be Type M (medium rate of absorption to the blood) for all radionuclides except ^{134}Cs and ^{137}Cs (Type F) and thorium (Type S). These selections are based on recommendations of the International Commission on Radiological Protection (ICRP 72) as cited in Eckerman 1999.

Probabilistic Evaluation of Dose and Risk

A probabilistic assessment was performed using the final soil guideline values as the initial soil concentrations to obtain estimates of the uncertainties associated with the dose and risk values calculated for the resident farmer. The assessment was performed as described above using distributions for the most sensitive parameters listed in ***Table D.2.*** and the area of contaminated zone.

Sludge Limits

The final soil guideline values were used to calculate sludge limits for each of the 12 radionuclides of interest based on the expected dilution (i.e., mixing of sludge into soil) and the amount of loss to radioactive decay during the land application period. For these calculations, residency is assumed to begin immediately following the last application of sludge. It is conservatively assumed that no radionuclides are lost via leaching or erosion during the land application period.

On a per acre basis, the total quantity of a key radionuclide that can be present on the land application site at the time residency begins is the soil guideline multiplied by the mass of the corresponding soil volume of 6.1 x 10^2 m^3 (assuming a 0.15 m mixing depth):

$$Q_a = C_{soil\ a} \times m_{soil}$$

$C_{soil\ a}$ = Concentrat ion limits for radionucli de "a" in soil, (pCi/g)

m_{soil} = Mass of soil per acre in top 15 cm (g)

Assuming a soil density of 1500 kg/m^3, the corresponding soil mass is 9.15 x 10^5 kg (9.15 x 10^8 g).

For a given radionuclide, the total activity that can be land applied annually on a per acre basis, assuming a constant input each year, without exceeding the corresponding soil guideline at year 20, can be calculated as follows:

$$I = \frac{Q_t \lambda}{(1 - e^{-\lambda t})}$$

I = Annual allowable input quantity (total activity) per acre (pCi/year)

Q_t = Quantity (total activity) per acre in top 15 cm at time, t (pCi)

λ = decay constant (years^{-1})

t = time (20 years)

The calculated annual allowable input quantity (total activity) can then be converted to a sludge limit by dividing the quantity by the mass of biosolids that are land applied on a per acre basis each year (4 dry tons/acre/year or 3.63 x 10^6 g assumed):

$$SL_a = \frac{I}{m_{sludge}}$$

SL_a = Sludge limit for radionucli de "a"

I = Annual allowable input quantity

m_{sludge} = Mass of sludge land applied annually (g)

RESULTS

The RESRAD calculated soil guidelines and corresponding sludge limits—based on a 10 mrem/year basic dose limit— for the 12 radionuclides of interest are presented in ***Table D.3.*** The ***Table D.3.*** values are single radionuclide guidelines and limits. When more than one radionuclide is present, a sum-of-the-ratios approach must be applied to demonstrate compliance with the single radionuclide soil guidelines and sludge limits. This approach ensures that the combined annual risk for all of the key radionuclides actually present does not exceed 10^{-4}.

Table D.3. RESRAD Calculated Soil Guidelines and Sludge Limits for Radionuclides of Interest.

Radionuclide	Soil Guideline[a] (pCi/g)	Sludge Limit ($pCi/g_{-dry\ wt.}$)
^{54}Mn	5.4	1100
^{60}Co	1.3	45
^{65}Zn	3.5[b]	900
^{90}Sr	3.2[c]	50
^{134}Cs	2.3	190
^{137}Cs	5.2	80
^{152}Eu	2.8	60
^{154}Eu	2.6	50
^{155}Eu	99	9600
Total U-Natural[d]	95 (46, 2, 47)[e]	1100
Total U-Depleted[d]	92 (77, 1, 14)[e]	1100
Total U-Enriched (1 to 3%)[d]	99 (16, 3, 79)[e]	1200

[a]Soil guidelines and sludge limits are calculated for single radionuclides (i.e., as if that were the only radionuclide present). For mixtures of radionuclides the sum-of-the-ratios must be calculated to determine compliance.

[b]Reduced from RESRAD calculated value of 39 pCi/g to ensure maximum risk estimate of 10^{-4}.

[c]A more conservative plant transfer factor of 0.95 was used for ^{90}S (RESRAD default = 0.3).

[d]The RESRAD calculated dose source ratios (mrem/year per pCi/g) for ^{238}U, ^{235}U, ^{234}U were used to calculate the total uranium values.

[e]Approximate activities of the individual uranium isotopes in the order ^{238}U, ^{235}U, and ^{234}U.

Risk Comparison

RESRAD 6.0 was used to estimate the risk of excess cancer associated with the radionuclides of interest at the calculated soil guidelines for the hypothetical resident farmer who establishes residency immediately following the last application of sludge. The risks associated with the previous soil guidelines (***Table D.1.***)—based on a 4 mrem/year dose limit—were also estimated for the radionuclides of interest and are presented in ***Table D.4.*** with the risk estimates for the final soil guidelines calculated for this work (based on 10 mrem).

Table D.4. Comparison of Previous Soil Guidelines and Corresponding Risks (based on 4 mrem/year) to Updated Values Based on 10 mrem/year				
Radionuclide	"4-mrem" Soil Guideline (pCi/g)	"10 mrem" Soil Guideline (pCi/g)	"4-mrem" Morbidity Risk	"10-mrem" Morbidity Risk
^{54}Mn	a	5.4	a	3×10^{-4}
^{60}Co	0.49	1.3	9×10^{-5}	2×10^{-4}
^{65}Zn	a	3.5	a	2×10^{-4}
^{90}Sr	a	3.2	a	1×10^{-4}
^{134}Cs	a	2.3	a	3×10^{-4}
^{137}Cs	2.0	5.2	7×10^{-6}	2×10^{-5}
^{152}Eu	1.1	2.8	9×10^{-5}	2×10^{-4}
^{154}Eu	a	2.6	a	2×10^{-4}
^{155}Eu	1.0	99	2×10^{-6}	2×10^{-4}
^{234}U	31	240	7×10^{-5}	8×10^{-5}
^{235}U	7.2	22	6×10^{-5}	2×10^{-4}
^{238}U	21	92	6×10^{-6}	3×10^{-5}

[a]This radionuclide was not addressed in the previous EA.

[b]Risk of experiencing a radiogenic cancer as a result of intake of the radionuclide or external exposure to its emitted radiations.

Uncertainty Analysis Results

The minimum, average, and maximum annual dose and risk estimates for the hypothetical resident farmer during the first year of residency (i.e., beginning immediately after the last application of sludge) are provided in ***Table D.5.*** It should be noted that the maximum values represent estimates in excess of the 95th percentile. For example, the maximum dose and risk estimates for ^{90}Sr are 41 mrem/year and 6×10^{-4}, respectively, but the 95th percentile values are 13 mrem/year and 2×10^{-4}.

The uncertainty analysis results indicate that the ***Table D.3.*** soil guidelines are adequately conservative for use in managing radionuclide levels on the land application sites.

Table D.5. Results of Uncertainty Analysis for Radionuclides of Interest at "10 mrem" Soil Guideline Concentrations.

Radionuclide	Statistic	Dose at time = 0	Risk at time = 0
^{54}Mn	min	1.3	2×10^{-4}
	max	12	4×10^{-4}
	avg.	6.5	2×10^{-4}
^{60}Co	min	0.70	1×10^{-4}
	max	12	3×10^{-4}
	avg.	6.7	2×10^{-4}
^{65}Zn	min	0.38	9×10^{-5}
	max	37	9×10^{-4}
	avg.	18	1×10^{-4}
^{134}Cs	min	5.1	1×10^{-4}
	max	14	3×10^{-4}
	avg.	7.3	1×10^{-4}
^{137}Cs	min	2.3	7×10^{-6}
	max	14	2×10^{-4}
	avg.	7.6	3×10^{-5}
^{90}Sr	min	0.19	6×10^{-6}
	max	41	6×10^{-4}
	avg.	3.7	6×10^{-5}
^{152}Eu	min	4.6	1×10^{-4a}
	max	11.7	3×10^{-4a}
	avg.	6.8	2×10^{-4a}
^{154}Eu	min	4.6	1×10^{-4}
	max	12	3×10^{-4}
	avg.	6.8	2×10^{-4}
^{155}Eu	min	4.6	1×10^{-4}
	max	12	3×10^{-4}
	avg.	6.8	2×10^{-4}
^{235}U	min	0.64	1×10^{-4}
	max	12	2×10^{-4}

Table D.5. Results of Uncertainty Analysis for Radionuclides of Interest at "10 mrem" Soil Guideline Concentrations *(Continued)*.

Radionuclide	Statistic	Dose at time = 0	Risk at time = 0
^{235}U (cont.)	avg.	6.5	1×10^{-4}
^{238}U	min	2.5	2×10^{-4}
	max	35	3×10^{-4}
	avg.	6.7	6×10^{-5}
^{234}U	min	0.75	2×10^{-5}
	max	16	1×10^{-4}
	avg.	7.1	3×10^{-5}

[a]Values are hand calculated because of an apparent error in the RESRAD results.

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